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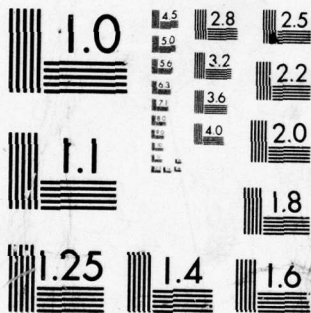
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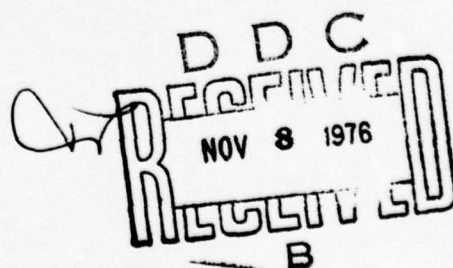
PECULIARITIES IN AITKEN NUCLEI COUNTS IN THE
TROPICAL AND SUBTROPICAL STRATOSPHERE

Josef Podzimek (GCCPR-UMR, Rolla)
W. A. Sedlacek (LASL-UCLA, Los Alamos)
J. B. Haberl (GE-Pittsfield)

Final Report to the ONR, Contract N00014-75-C-0413 ✓

and to the

U. S. Department of Transportation, CIAP



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PECULIARITIES IN AITKEN NUCLEI COUNTS IN THE
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INTRODUCTION

March 1974 saw the start of aircraft measurements with the new General Electric Stratospheric Aitken Nuclei Detection System (SANDS). These measurements were a part of the broad Climatic Impact Assessment Program (CIAP) of the U.S. Department of Transportation. One of the main goals was to supply data about the background concentrations of stratospheric aerosols before the SST aircraft begin to operate in higher altitudes.

During the series of measurements performed in 1974 and 1975 over New Mexico, Texas, and the Rocky Mountains, Aitken nuclei (AN) concentrations ranged from several hundreds or thousands per cm^3 (around the tropopause level) down to several tens of nuclei per cm^3 at the altitude of 18 km (Podzimek *et al.*, 1975). Higher nuclei counts and an intense fluctuation of nuclei concentration were detected above the ridges of high mountains during meteorological situations characterized by well developed jet-stream in the troposp here.

Both series of flights in November 1974 and February 1975 indicated a dramatic change in AN concentration which was accompanied by a change in ozone concentration. The purpose of this article is to analyze these changes

and to find a possible explanation of the peculiarities in AN counts in the tropical and subtropical stratosphere.

FLIGHTS IN NOVEMBER 1974 AND FEBRUARY 1975

The SANDS instrument and its calibration were described elsewhere (Haberl, 1975; Wegrzyn and Podzimek, 1975). A survey of the flights and AN measurements performed in 1974 and 1975 was published in special reports (Podzimek *et al.*, 1974; Podzimek *et al.*, 1975). Since the SANDS provided credible results and since the AN counts apparently reflected the physical state of the lower stratosphere, this discussion will survey the following questions: What general characteristics of AN concentrations can be deduced from the flights in the subtropical and tropical stratosphere?; what are the possible explanations and interpretations of the observed AN concentration peculiarities?

a) Vertical Ascents

During the time period mentioned there were four vertical aircraft ascents, the first of which (November 18, 1974) was partly described in a previous report (Podzimek *et al.*, 1975).

November 18, 1974; 20:50 to 21:15 hrs. GMT; Houston

The flight was characterized by AN counts ranging from 60 particles per cm^3 at an altitude of 19.0 km to 700 particles per cm^3 at 7.5 km. AN concentrations were not strongly stratified with altitude and the mean

concentrations were not much different during the ascent and descent of the aircraft.

The negative correlation between AN counts and ozone concentration at altitudes above 17 km is conspicuous. Ozone concentration (Fig. 1) was measured by the instrument of Dr. Rosen from the University of Wyoming and is plotted in relative units because the final calibration was not available. 100 units in Fig. 1 correspond roughly to 350 nanobars of partial ozone pressure. At lower levels the ozone seems to follow the profile of AN concentration around 15 km. However, the positive correlation is not perfect, which was apparent in several layers above 10 km which could be due to the influence of the tropical or middle-latitude tropopause.

February 11, 1975; 14:19 to 15:39 hrs. GMT; Houston

AN concentrations (Fig. 2) had almost the same profile between 7 and 14 km during the ascent and descent of the aircraft. There were high counts around the altitude of 7 km (700 AN cm^{-3}), a step of higher concentrations below the tropopause (over 200 AN cm^{-3}), and a very steep decrease of AN above the tropopause to 60 AN cm^{-3} at 13.5 km.

Ozone concentrations show a negative correlation with AN above the tropopause and a positive correlation below it. However, the flight altitude was too low and the ozone concentrations at 14 km did not seem to decrease proportionally to the increasing AN counts (from 60 AN cm^{-3} at 13.5 km to 90 AN cm^{-3} at 14.0 km).

February 15, 1975; 18:02 to 19:18 hrs. GMT; Panama

On this specific day two ascents were made with the instrumented aircraft within three hours (Fig. 3). This was a good check on the reproducibility of AN measurements. Each point represents the mean value of 150 individual measurements and sometimes there were several measurements at one level. A closer look at each point reveals that the deviations of individual measurements are insignificant and that the reproducibility of AN measurements is very good if one bears in mind the fact that the ascent measurements were not performed in exactly the same place but over the canal zone and southward of it. The most interesting phenomena of the AN vertical profiles in tropical regions is the smooth curve of nuclei concentrations up to the tropopause level. Almost the same AN concentrations existed between the altitudes of 6 and 12 km (around 300 AN cm^{-3}). Then followed a drop in counts, below the tropopause, ranging around 130 AN cm^{-3} with a continued slight decrease of AN concentrations up to the 19 km level.

There is strong support of the findings in Fig. 1 that a negative correlation between AN concentration and ozone exists above the tropopause and (usually) a positive one below it. The differences in ozone concentrations during ascent and descent which were measured between 13 and 16.5 km cannot be explained at this time.

February 21, 1975; 16:22 to 16:49 hrs. GMT; Houston

The main features of the AN concentration vertical profile (Fig. 4)

are the following: The nuclei concentrations ranged between 620 AN cm^{-3} at 9 km down to 50 AN cm^{-3} at 15 km with a markable increase in AN counts at 9 km and 13.5 km. There is a certain analogy between the vertical ascents above Houston on November 18, 1974 (Fig. 1), and February 11, 1975, and this particular ascent. In a layer between 6 and 9 km, and a layer between 13 and 15 km, there was an apparent increase in AN concentration.

Ozone concentrations in the troposphere were not related to the AN counts and the indication of a negative correlation with AN counts in altitudes above the tropopause was not very reliable because the airplane did not take the samples at the same place. Also, the flight altitude (maximal at 15.5 km) was not sufficient for obtaining the negative correlation which was evident during the flights on November 18, 1974, and on February 15, 1975.

b) Horizontal Flights (Fig. 5)

November 21, 1974; 10:27 to 12:43 hrs. GMT; from Houston to the south over
over the Gulf of Mexico (Fig. 6)

This flight was partly described in the previous report. The aircraft climbed over the Houston area and then followed a southward course at an altitude of 18.5 km. AN counts were below 100 particles per cm^3 along the path until 26°N latitude. There the concentration started to fluctuate between approximately 100 and 200 AN cm^{-3} and afterwards remained almost at a constant concentration of 200 AN cm^{-3} . There were no substantial differences between the counts before and after the sunrise (12:16 hrs. GMT) at this altitude.

Ozone concentration, measured with the instrument of Dr. Rosen from the University of Wyoming, had an opposite trend to AN. This negative correlation was apparent even during the fluctuating AN concentrations between 26°N and 22°N latitude.

A qualitative comparison of size spectra of AN measured northward of 26°N latitude (described in more detail in the previous report) showed a substantial difference between the size spectra measured on November 15, 18, and 21 at different altitudes. Although the measurements of the AN spectrum were based on several questionable assumptions (validity of Boltzmann's distribution law, constant ratio between charged and uncharged particles, laminar flow in the cylinder during the fast operation of the counter) one can draw a very important conclusion from these measurements. The AN composition on November 21, 1974 at 18.2 km prior to reaching the 26°N latitude was different from the size spectrum taken at that latitude. The latter was characterized by two distinct groups of particles with radii smaller than $0.02\ \mu\text{m}$ and larger than $0.05\ \mu\text{m}$.

February 13, 1975; 14:48 to 19:37 hrs. GMT; from Houston to Ecuador (Fig. 7)

The flight was performed almost entirely at a level of 18 km after climbing up southward of Houston. AN counts were low, varying between 20 and $40\ \text{AN cm}^{-3}$. Just before flight altitude was reached (at 27°N latitude), there were more intense fluctuations of AN counts accompanied by conversely fluctuating ozone concentrations. The ozone concentrations, however, were

disturbed due to the fact that the aircraft was still climbing. A little stronger fluctuations of AN concentrations were found northward of Yucatan and Honduras and also above 7° N latitude. The latter case could possibly be related to the more intense exchange above high mountains or more likely to the relative proximity of the Intertropical Convergence Zone (ITCZ). There were no changes in AN counts above the equator at 18 km.

Ozone concentrations were almost constant during the whole flight. Above the equator they amounted to 5 (in relative units) and northward of 20° N latitude they were slightly higher.

February 14, 1975; 19:53 to 22:02 hrs. GMT; southward of Panama to Peru

(Fig. 8)

The only part of the flight which yielded usable AN data was approximately at the level of 16.5 km. The aircraft flew from the point 4° N, 79° 36'W to 9° 34'S, 78° 28'W (Peru). AN concentrations were, in mean, higher and the fluctuations of counts were more intense than on the previous day. The nuclei concentrations were between 35 and 80 AN cm^{-3} above the equatorial region. However, southward of 5° S latitude, the fluctuations increased their amplitudes and several times they surpassed 100 AN cm^{-3} . This interesting change in AN concentration horizontal profile around 6° S latitude was not accompanied by any dramatic change in ozone concentration. There is a slight indication of a negative correlation above 2° N and 8° S latitude. Ozone concentrations varied between 2 and 12 (in relative units) and were very stable.

February 15, 1975; 19:23 to 21:47 hrs. GMT; southward of Panama to Ecuador and back (Fig. 9)

The main purpose of this flight was to study the AN concentrations above the ITCZ and above the equator. For this reason, the flight was performed at two different levels: above the altitude of 18 km toward the equator and around 15 km on the flight back. The flight toward the equator was characterized by very low AN counts. The concentrations varied between 12 and 20 AN cm⁻³ and were very stable. The flight back, which was performed at a lower altitude, presented quite a different profile of AN concentrations. Strong fluctuations of counts were characteristic for the flight above the continent with counts often surpassing 150 AN cm⁻³. Maximal concentrations were found above the equator (more than 180 AN cm⁻³) and above the supposed location of the ITCZ (more than 150 AN cm⁻³ around 5° N latitude).

Ozone concentration at higher altitudes was very steady with the maximum corresponding to the highest flight altitude (11 in relative units) and lowest above the equator (6.5). At an altitude of 15 km, the ozone concentration profile apparently was not related to AN.

February 21, 1975; 16:22 to 22:28 hrs. GMT; from Houston northward to the Canadian frontier (Fig. 10)

The flight northward was performed around the altitude of 15 km and the flight back (with a gap in data of more than half an hour) around the level of 18 km. The lower level flight yielded AN counts varying between 13 AN cm⁻³

(on the northernmost points of the path) to more than 50 AN cm^{-3} above the Houston area. There were strong fluctuations of AN concentrations along the whole path showing no correlation with the corresponding ozone counts. The flight back was characterized by slightly higher counts on the northernmost part of the path (around 15 AN cm^{-3}) and lower counts above Houston (around 10 AN cm^{-3}). There is indication that the low AN counts between 38° N and 45° N latitude correspond to the maximum in ozone concentration (above 70 relative units).

In general, ozone concentration was higher above the northernmost point at 18 km (50 relative units) than above the Houston area at the same level (20 relative units). The ozone counts were, however, much higher than during the flights over tropical regions.

PECULIARITIES OF NOVEMBER 1974 AND FEBRUARY 1975 FLIGHTS

From the four vertical and five horizontal flights it seems appropriate at this time to extract some typical values from the variety of geographical locations and environmental conditions. Only some possible reasons for variations will be discussed rather than the formulation of formal statements of mean concentration profiles. Due to the limited number of flights which were performed mainly in the winter season the latter would be, at best, a statistical abstraction.

Vertical profiles over Houston were characterized by a layer of higher AN counts usually located between 7 and 9 km ($600\text{--}800 \text{ AN cm}^{-3}$) and by

increased AN concentration just below the middle-latitude tropopause (around 12 km). The tropical tropopause and the stable air stratification above it probably causes another increase in AN concentration at altitudes higher than 15 km (clearly seen in Fig. 1). This high altitude maximum in AN concentration (250 AN cm^{-3} in Fig. 1) is comparable with the increase in AN counts below the middle latitude tropopause. There is a basic difference between the AN vertical profiles measured above the Houston area on November 18, 1974 and on February 11 and 21, 1975. The November 18 measurement shows a strongly expressed layer type structure and also greater differences between ascent and descent AN concentrations. Considering the fact that each point on the AN concentration curve represents more than 100 individual measurements, the differing nature of both of the profiles obtained in February is apparent. The lower level strata of higher AN concentration might be related to air traffic or to pollution of the troposphere from sources located on the ground. The high level variations in AN counts can be caused by world wide sources of stratospheric pollution (Guatemalan volcano Fuego eruption during the flight on November 18, 1974; McCormick and Fuller, 1974), or by air traffic over high ridges of mountains (Podzimek et al., 1975; Cadle and Langer, 1975). Other possible causes of variations are the exchange of air masses around jet streams and thunderstorms (Cadle et al., 1969; Weickmann and Van Valin, 1972), and stratospheric air circulation combined with thermal stratification of the atmosphere.

It is interesting to compare the AN vertical concentration profiles over Houston (this report) and Albuquerque (Podzimek *et al.*, 1975) with the profiles measured in West Germany 53° N) by Kaselau *et al.*, 1974. There is no difference in the shape of the curves or in the mean AN concentrations. The October 1973 balloon ascents yielded concentrations around 33 AN cm^{-3} between 5 and 10 km with a markable increase just below the tropopause. At higher levels, the AN concentration dropped steeply to a value of 20 AN cm^{-3} . Several balloon flights with AN counter similar to that of Junge's showed no substantial changes in AN concentration above the 20 km level ($10 \text{ to } 20 \text{ AN cm}^{-3}$). These results and also AN balloon measurements in Wyoming show that above Junge's layer, one might anticipate a source of AN to keep their counts relatively constant at least up to the level of 27 km.

Vertical profiles over Panama (ascent and descent measurements are plotted in Fig. 3) showed almost a constant AN concentration up to the level of 12 km (around 300 AN cm^{-3}). Above this there was a steep decrease in counts followed by a small increase just below the tropical tropopause. AN concentrations above 18 km were lower than at the same altitude above Houston ($10 \text{ to } 20 \text{ cm}^{-3}$ compared to $60 \text{ to } 90 \text{ cm}^{-3}$).

Ozone concentrations were apparently negatively correlated with AN counts in high altitudes (above 17 km). In the measurements over Panama, one can see this negative correlation just above the tropical tropopause. The lower levels over the middle latitudes, including the layer above the tropopause

show no clear correlation between ozone and AN counts. However, over Panama and the ascent over the Houston area (February 11, 1975) showed a slight indication of a positive correlation of both parameters in the troposphere above 6 km altitude.

Horizontal flights covered the latitudinal distance from almost 10° S to the Canadian frontier (48° N). There were some interesting features of the flights over tropical, subtropical, and equatorial regions. Both flights (November 21, 1974; February 13, 1975) yielded higher AN counts around the latitude of 27° N with a sudden variation of AN counts afterwards. This AN concentration increase was accompanied by a decrease in ozone concentration. Because both flights were performed within the time span of several months, one can (in the authors' opinion) exclude the influence of the activity of the volcano Fuego which was hypothesized earlier (Podzimek *et al.*, 1975). The altitude at which it happened (18 km) and the wave like course of the concentrations strongly indicate that the aircraft entered the tropical tropopause and that it is either folded or layer type structured. A rough estimate of the first wavelength fluctuation of AN and O_3 concentration was 60 miles (February 13) and 70 miles (November 21).

The qualitative change in AN size spectrum assumed earlier (Podzimek *et al.*, 1975) can be explained by the exchange or aging of larger particulates in the troposphere. If one accepts the hypothesis (Blifford, 1970) "that the

upper tropopause contains aged particles whose distribution has been modified by physical and chemical processes", then this explains the "plateau" in the size spectrum distribution curves found during the flight on November 18, 1974. This "plateau", revealing larger quantities of particulates with radii between 0.02 and 0.06 μm can be due to the chemical reactions of particulates with gaseous substances (i.e. Friend et al., 1973) or simply to the penetration of humid tropospheric air into the stratospheric levels and the subsequent condensation of water vapor on nuclei.

During the horizontal flight on February 13, 1975 at 18 km, there was very little if any indication of the Intertropical Convergence Zone (ITCZ) around 6°N latitude which could also be due to the influence of high mountains over the central part of Ecuador. Near the equator, the AN counts remained low (around 20 AN cm^{-3} at 18 km).

The flight on February 14, 1975 was performed to the west of the previous flight and at an altitude of around 17 km. The AN concentrations fluctuated around 50 AN cm^{-3} with a sudden and markable increase of AN counts over Peru. In this broad band of higher AN concentrations extending from 5°S to 9°S latitude the concentrations surpassed the value of 100 AN cm^{-3} several times. These higher counts might be related to the influence of high mountainous ridges of the Andes, or possibly to the ITCZ. The first assumption would, however, assume that the higher counts would start as soon as the aircraft came over the Andes. The second assumption is related to the open

question about the secondary ITCZ. The latter merits more attention due to the findings by Hubert et al. (1969) who analyzed the ESSA satellites' (number 3 and 5) cloud pictures. They found a strong support that in winter months (December to February), there is a temporarily expressed second ITCZ to the south of the equator which was crossed by the path of the aircraft on February 14, 1975. However, on the satellite cloud picture of February 14, 1975, there is only a part of the ITCZ suggested by Hubert et al., stretching from Peru to central Brazil. Its structure is not of the type of narrow bands of clouds seen over the Atlantic or Pacific north of the equator. The strong influence of high mountains is apparent in modifying the ideal shape of the ITCZ and giving origin to the convective clouds of high vertical extent which cover a large area of Andes.

There was a very low and uniform concentration of ozone during the flight path on February 14, 1975. Only small increases in ozone concentration were observed around 2° N and 8° S latitude. One might wonder just how high the disturbances caused by convection in the ITCZ penetrate into the troposphere. The aircraft did not fly above the tropopause on the February 14 flight.

The flight on February 15, 1975 from Panama toward the equator was intended to answer this question. The aircraft flew southward at an altitude higher than 18 km and back at approximately 15 km altitude following the same course. There was a conspicuous difference between both AN concentration

profiles. Above the tropopause there were very low counts of AN, usually below 20 AN cm^{-3} . No strong variations of AN counts were observed during the flight. The ozone concentrations followed approximately the altitude profile of the aircraft path reaching (in mean) 10 in relative units. On the flight back the aircraft passed through a zone of strong convection over the peaks of the Andes which caused an increase in AN counts. Measured concentrations in this zone often surpassed 180 AN cm^{-3} . Further to the north the aircraft flew over cloud clusters belonging to the ITCZ system, which one can easily follow in the satellite cloud picture (12:00 GMT). There is a close relationship between a mighty cloud cluster around 4° N latitude and the measured peak in AN concentration, and between the cloudless sky and the low and very uniform AN counts (100 AN cm^{-3}) northward of 5° N latitude.

Summarizing the finding on the level of penetrating disturbances from the troposphere, one can conclude that the tropical tropopause seems to almost completely suppress the vertical transport of AN. This transport, on the other hand, is strongly dependent upon the convective activity in lower levels and is seen in AN count records in higher tropospheric levels.

A second checking on the AN exchange in higher levels was made during the last flight of this series from Houston to the Canadian border and back on February 21, 1975. The first part of the flight was performed at an altitude of approximately 15 km. It was characterized by strong fluctuations of AN counts (between 13 and 64 AN cm^{-3}) and by low counts mainly in the

northern part of the flight. There were low AN concentrations at the low pressure center over the northern part of Missouri (ground chart) and at the location of the ridge in the 500 mb chart. The anticyclonic curvature of isolines still prevailed in the 200 mb chart, and it was also slightly indicated in the 100 mb chart. The flight back performed approximately at a level of 18 km did not show such dramatic fluctuations as the flight at 15 km. The AN counts were uniform and only in a few places did they surpass 20 AN cm^{-3} .

Unlike the AN counts, the ozone concentrations on the flight back at 18 km showed substantial changes ranging from 20 to almost 80 (in relative units). The highest ozone concentrations were found in the middle part of the record at 18 km in strong contrast with the almost uniform concentration measured at 15 km altitude. There was no negative correlation between AN counts and ozone at higher altitudes which was clearly found over tropical and subtropical regions. One of the possible explanations for this might be found in using the generally accepted model of air circulation in the lower stratosphere (i.e. Vincent, 1968). From November to March can be anticipated a downdraft in the stratosphere above the latitudes around 40° . This could explain the higher ozone concentrations in the middle latitudes which often have drastic changes in the lower stratosphere (Dutsch, 1974). Because the AN counts are very low in mean at an altitude of 18 km, and because unfortunately a part of the record is missing, it is impossible to find

a negative correlation with ozone which was so strongly expressed over the Gulf of Mexico. Nevertheless, evidence is convincing that such correlation exists mainly in the part of the record corresponding to the northern section of the flight.

CONCLUSIONS

↓ The analysis of AN concentrations measured during a series of flights over tropical and subtropical regions performed in the months of November 1974 and February 1975 gave the following conclusions/.

- 1) Vertical profiles of AN concentrations over Panama differ substantially from those over Houston. In winter months over Houston one can usually find strongly expressed layers of very high concentration AN in the troposphere. Often there was an increase of AN counts below the middle-latitude tropopause and below the extension of the tropical tropopause as well. AN vertical profiles over Panama have an almost constant AN concentration below the tropopause and a steep decrease of concentrations above it.
- 2) The mean AN concentration above equatorial regions is very low. At altitudes higher than 18 km it rarely amounts to over 20 AN^{cu}cm⁻³.
- 3) There is strong indication that convection in tropical regions correlates with AN counts when measurement is made below the tropopause. However, above the tropical tropopause there is no indication that AN are transported in a considerable concentration from the troposphere due to convection. → next page

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4) A negative correlation between AN and ozone concentration was found over the Gulf of Mexico at altitudes above 17 km. This correlation was not so strongly expressed over equatorial regions or north of 40^{deg} N latitude at the same flight level (18 km).

5) There was no apparent correlation between AN counts and ozone concentration just above the tropopause which was deduced by several authors for larger particulates, (i.e. Rosen et al., 1974).

6) A broad maximum in ozone concentration appears in February between 38^{deg} N and 44^{deg} N latitude which might be negatively correlated to AN counts. This statement, however, needs to be supported by more measurements in the future. ↗

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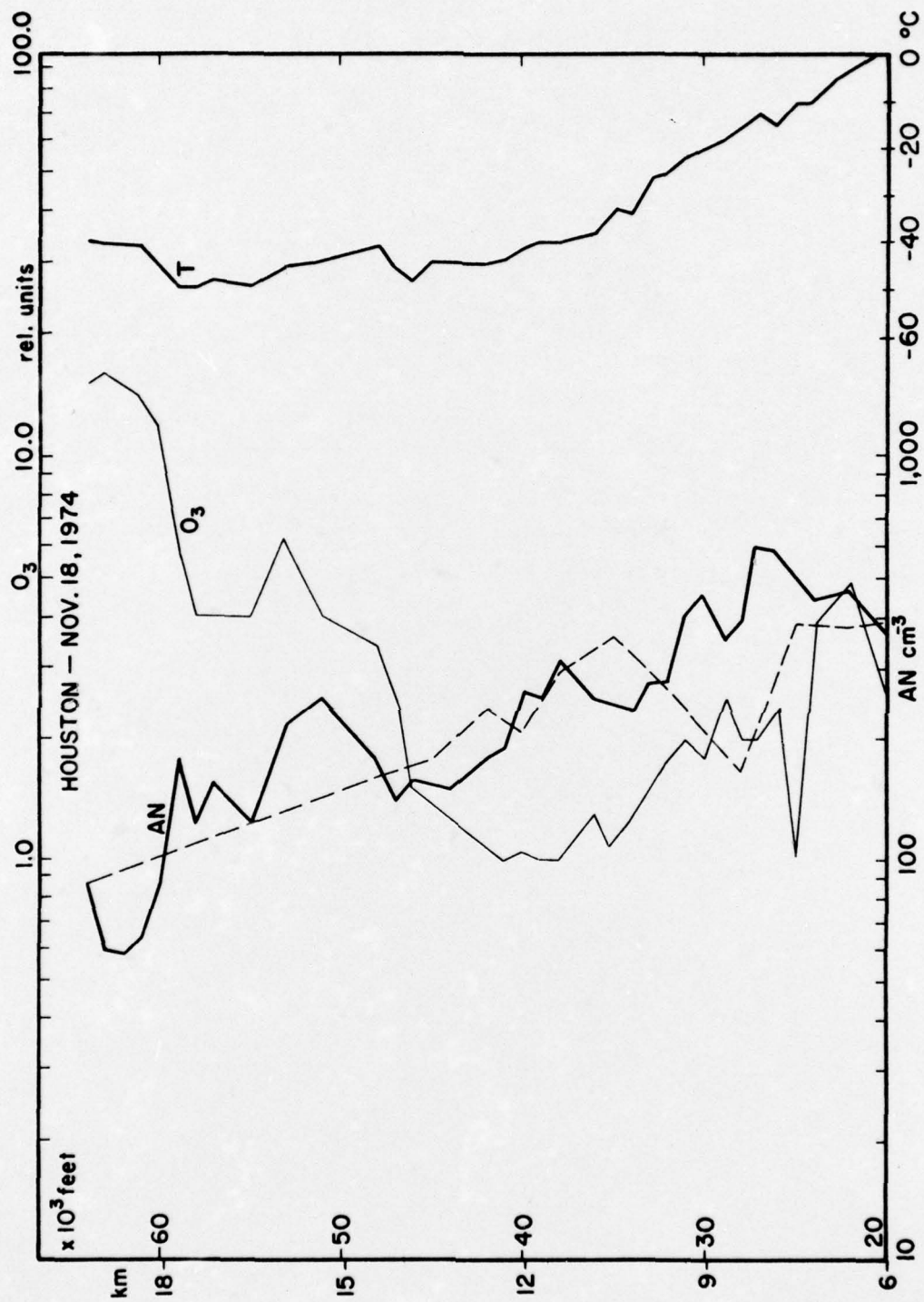


Fig. 1

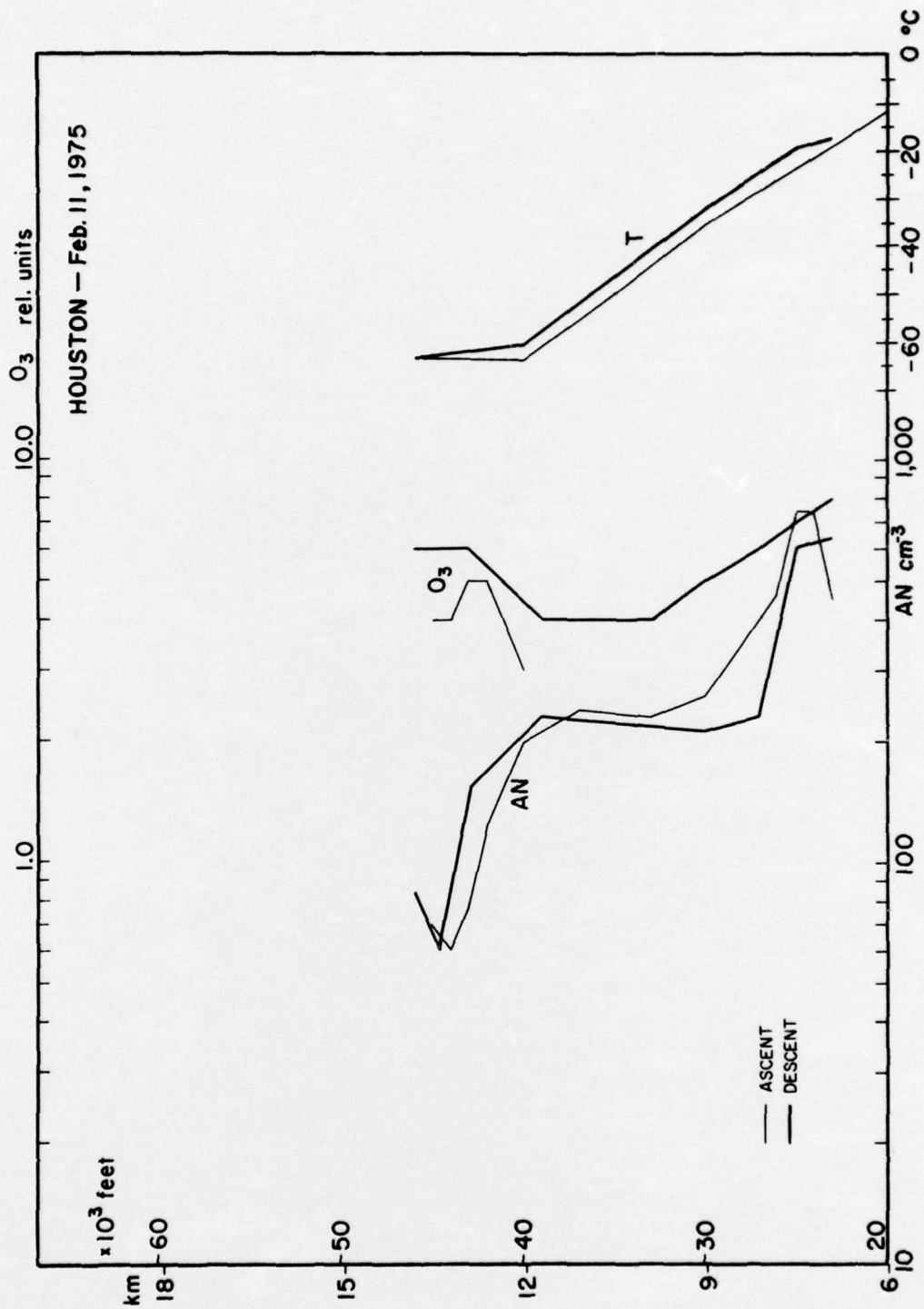


Fig. 2

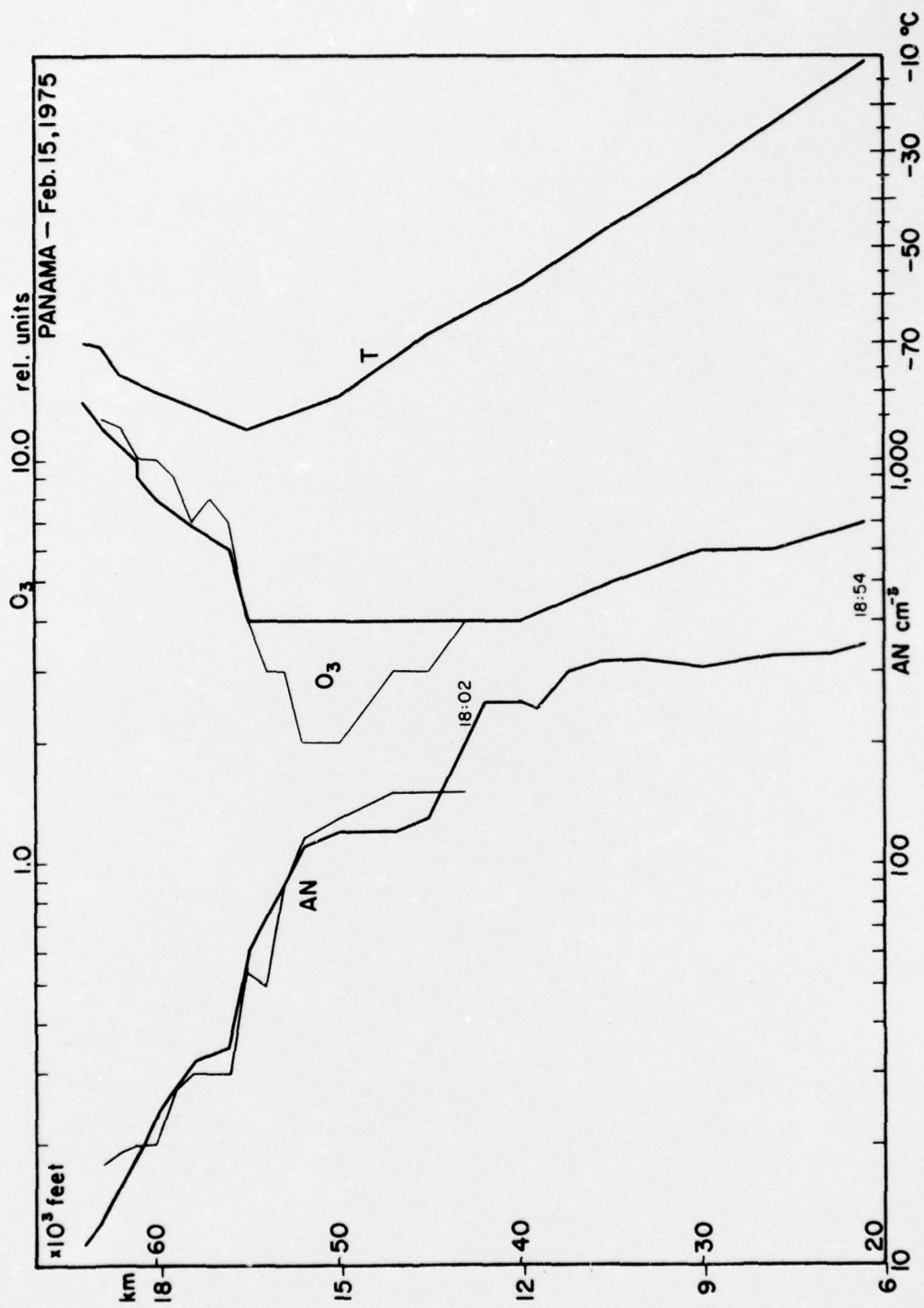


Fig. 3

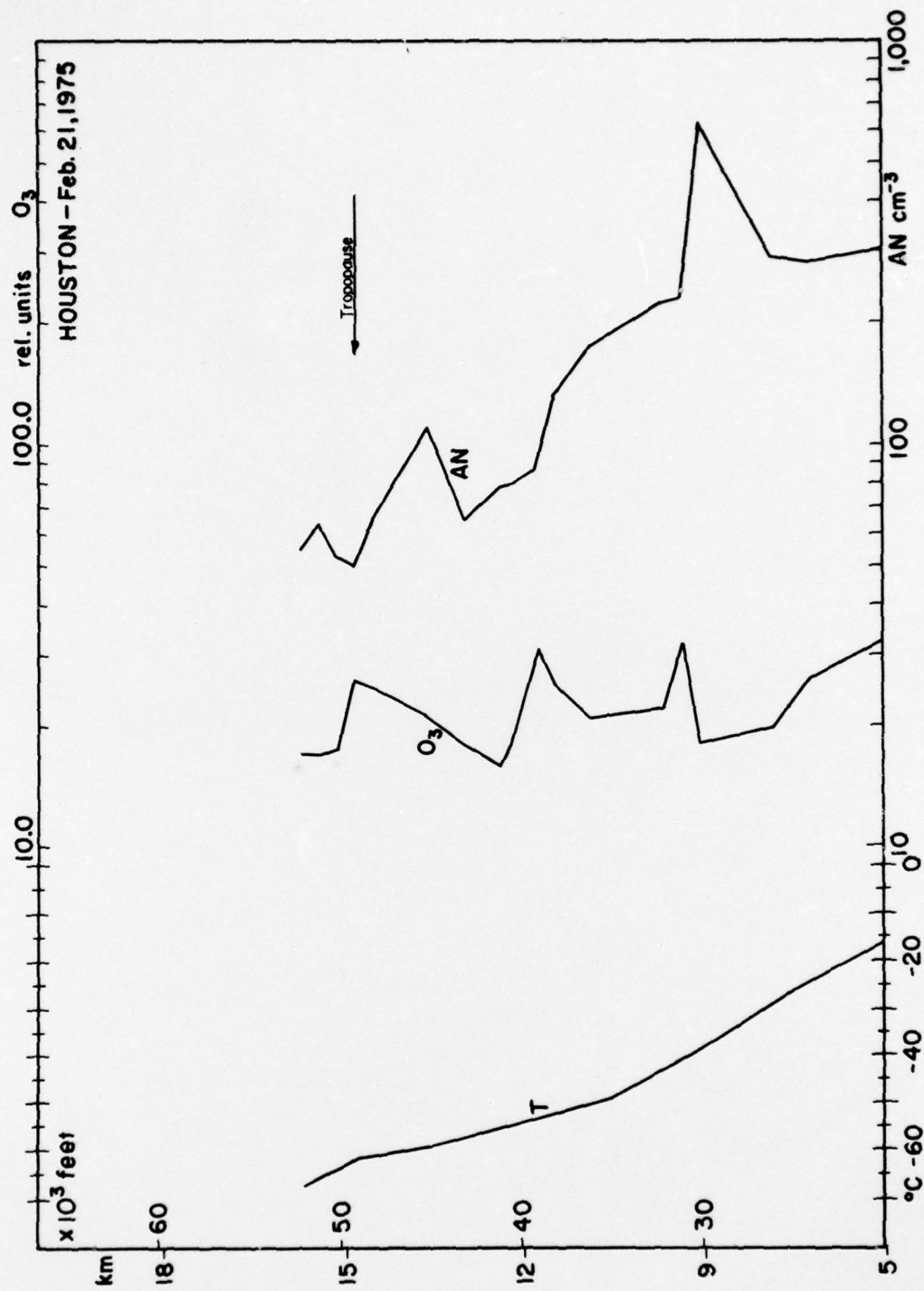


Fig. 4

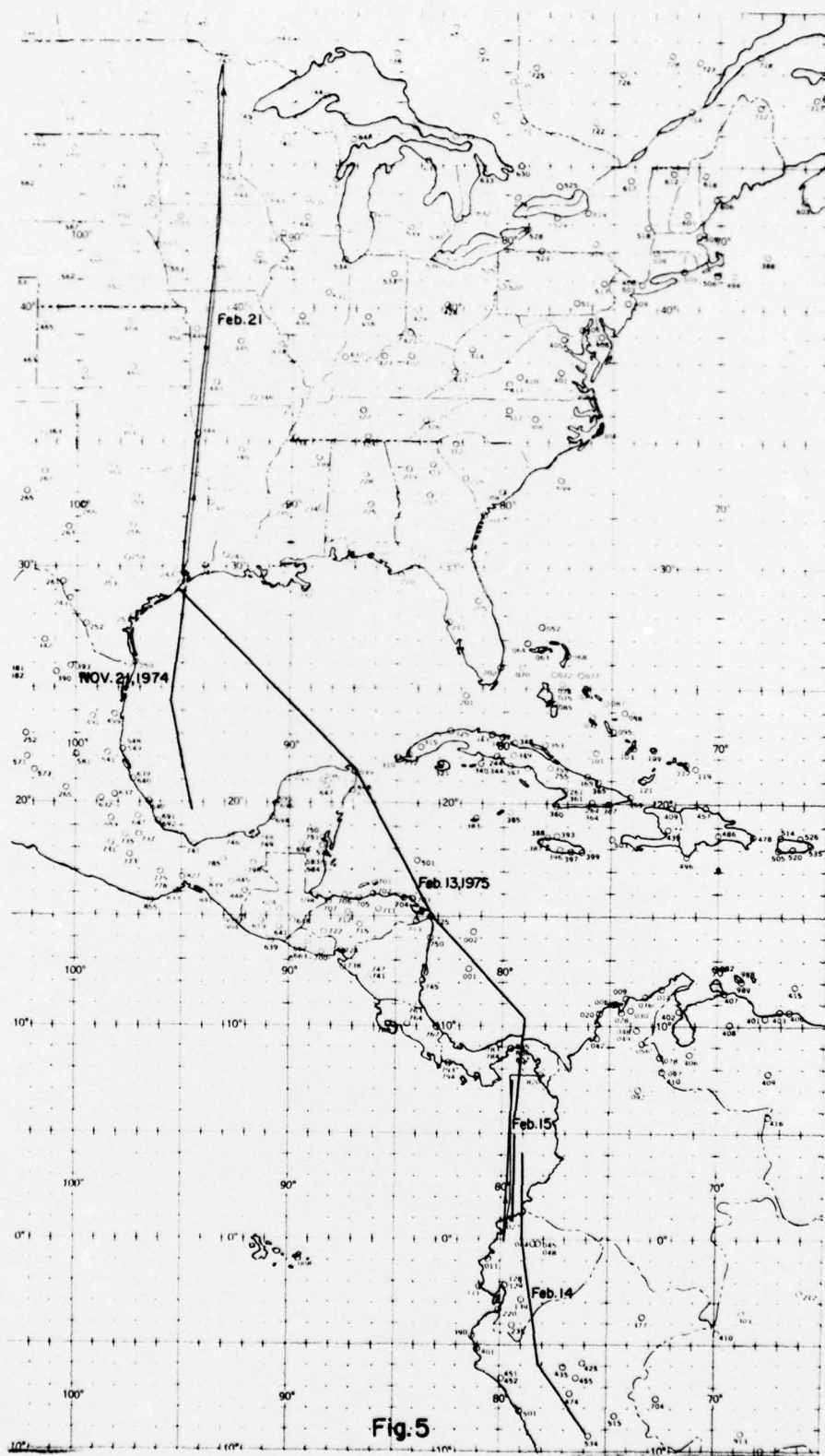


Fig. 5

12:16 HRS. GMT SUNRISE

11:43

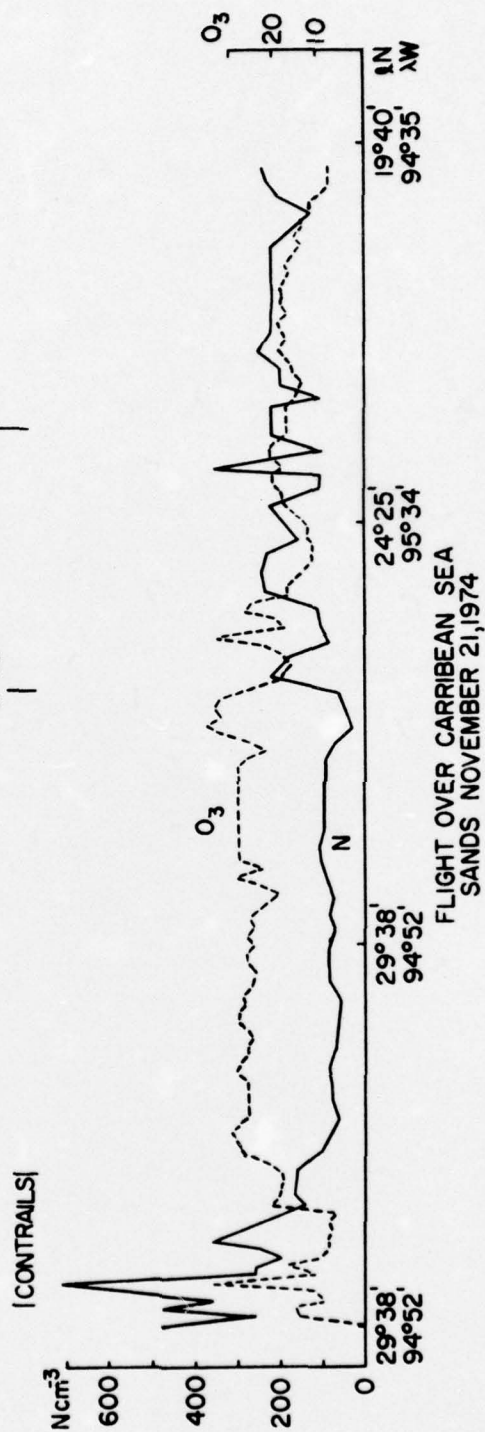
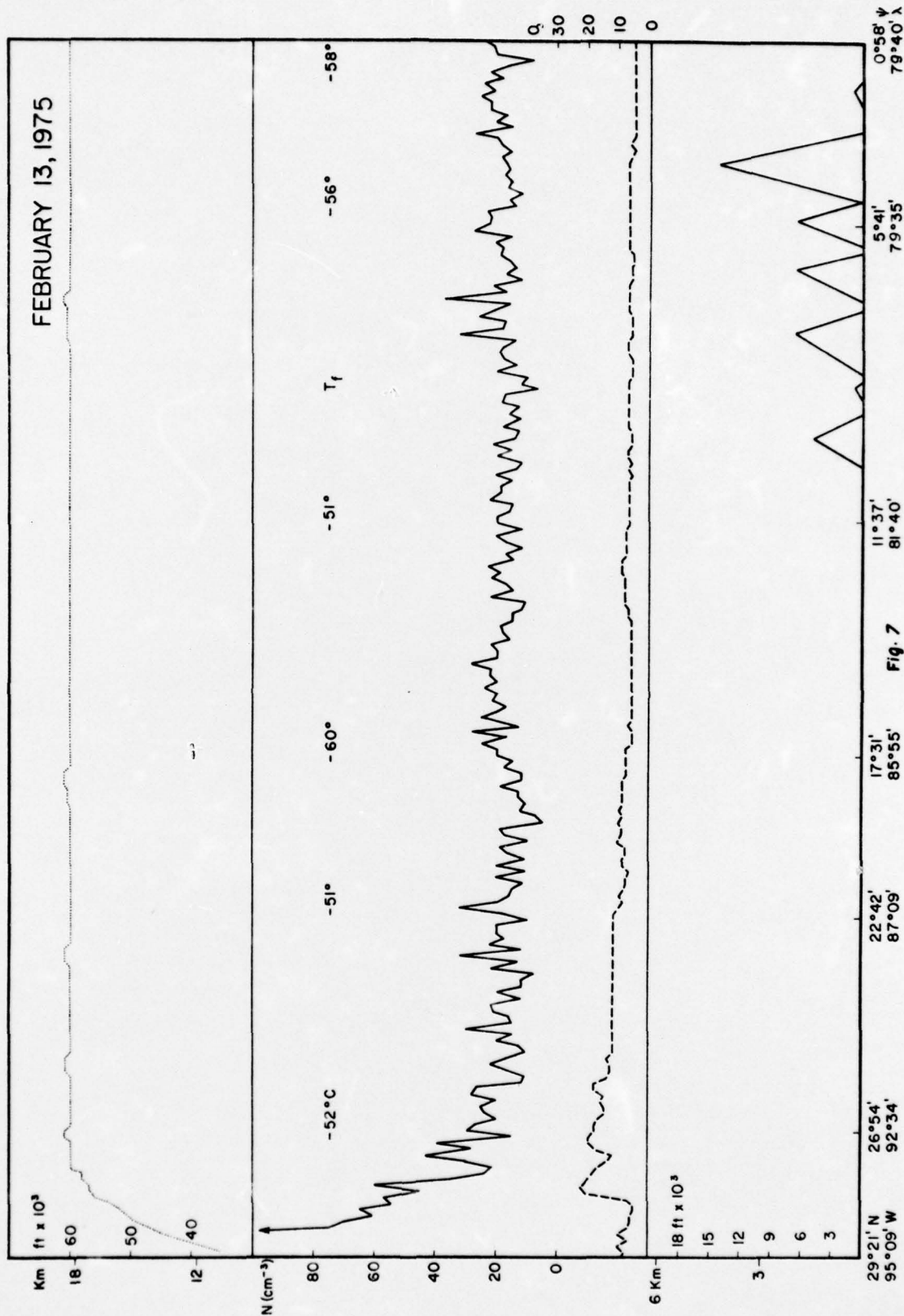


Fig. 6

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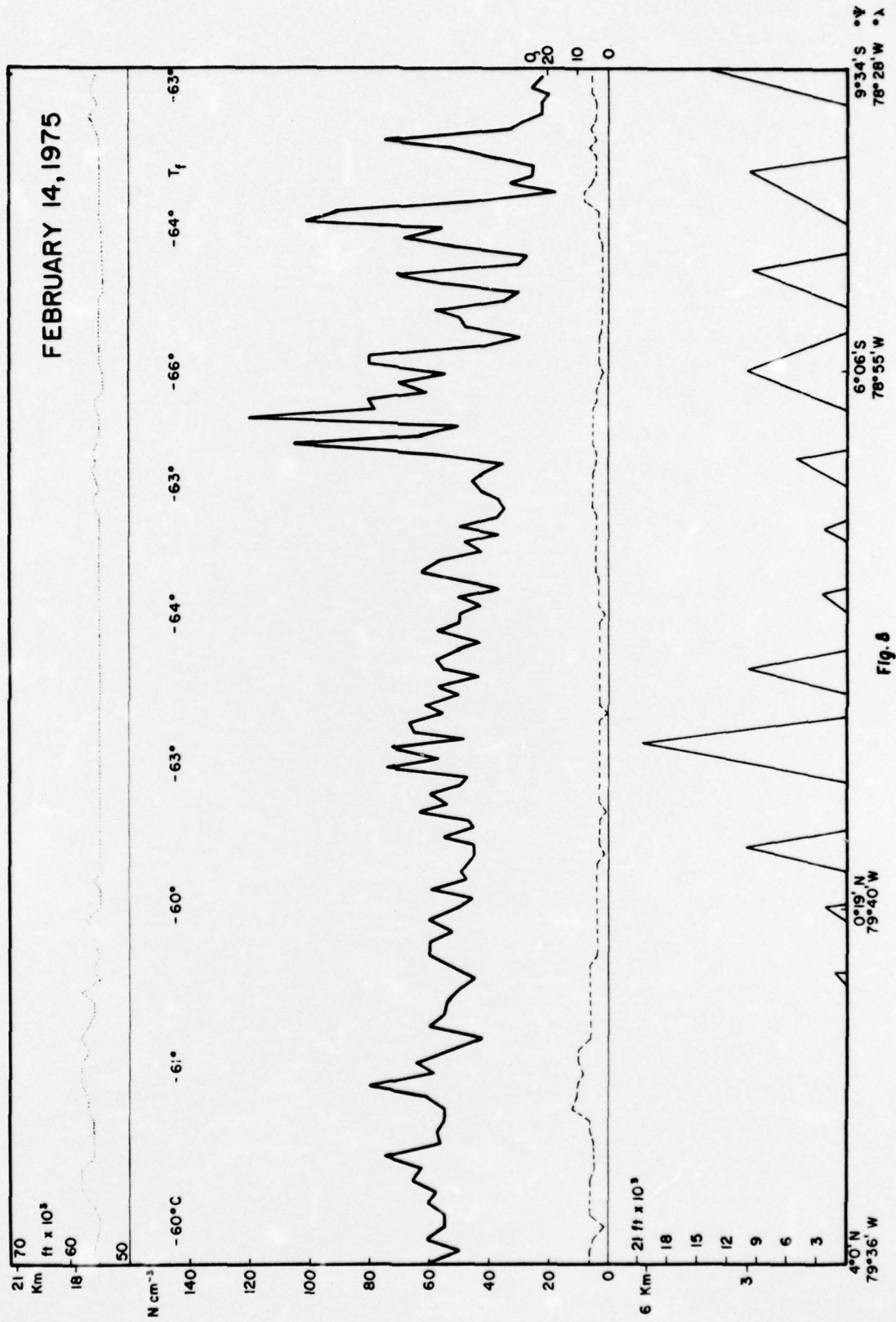


Fig. 8

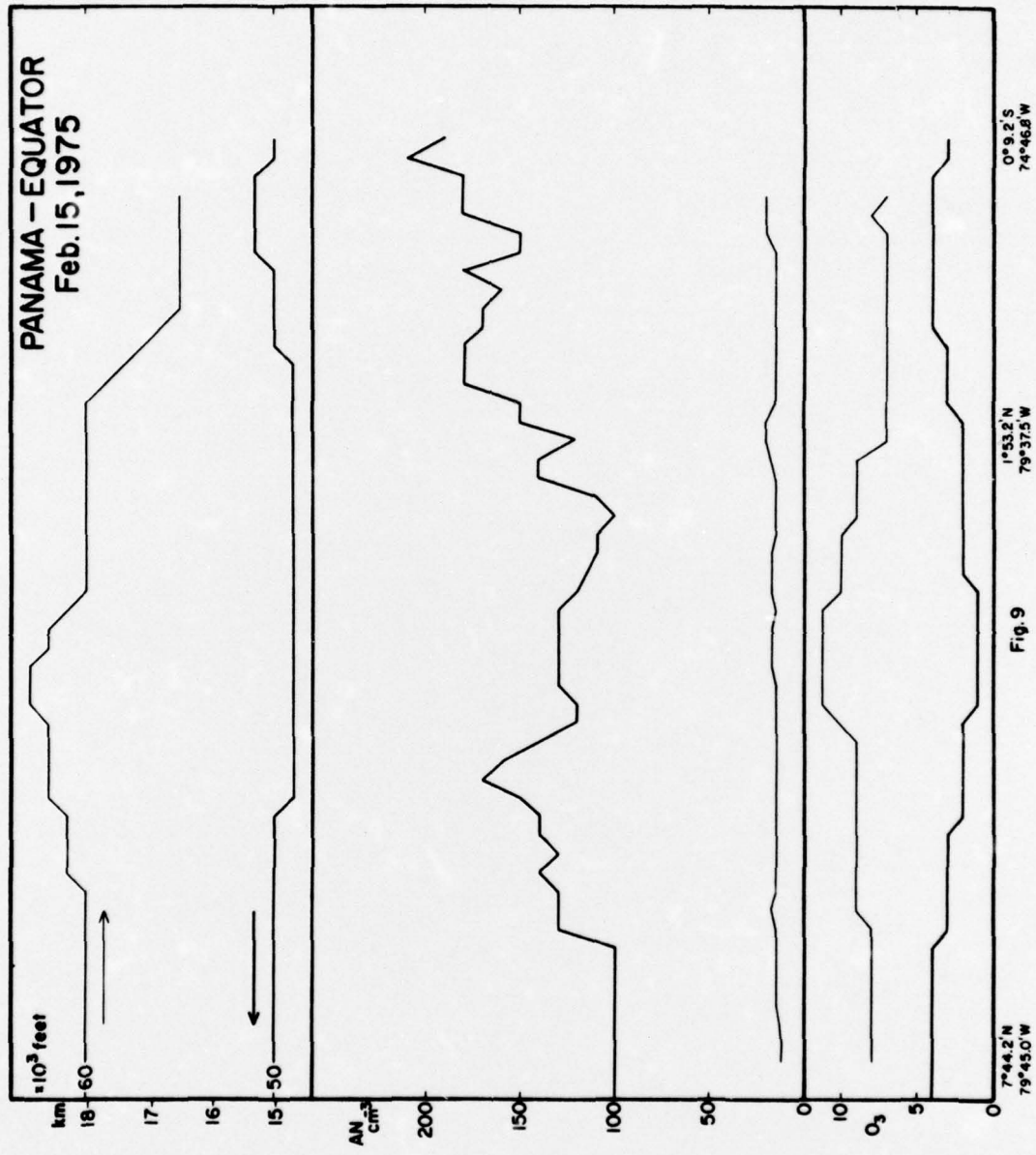


Fig. 9

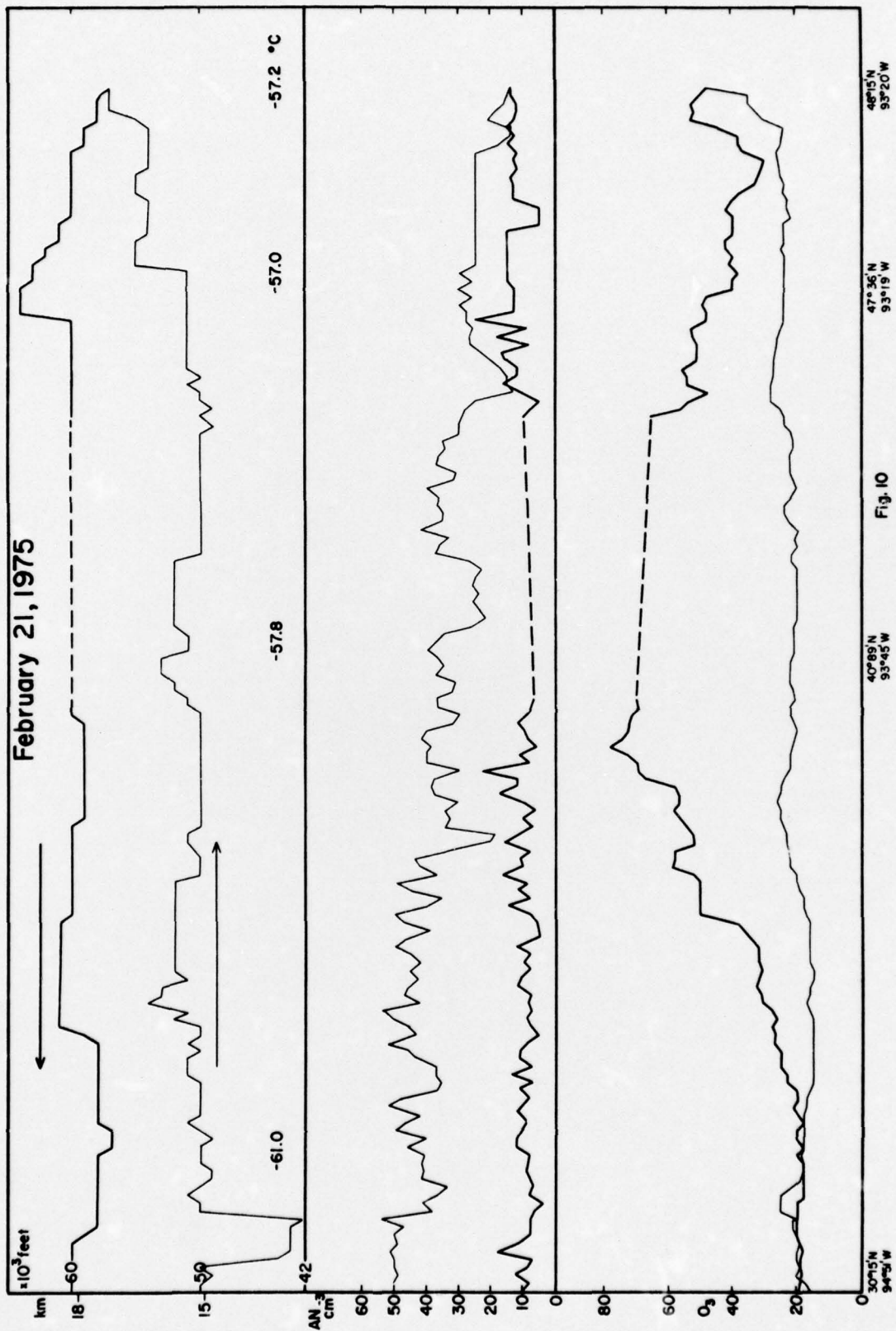


Fig. 10